Chinese Journal of Chemical Engineering, **16**(5) 663—669 (2008)

PERSPECTIVES

Gas-Liquid Microreaction Technology: Recent Developments and Future Challenges*

CHEN Guangwen (陈光文), YUE Jun (乐军) and YUAN Quan (袁权)**
Dalian Institute of Chemical Physics, Chinese Academy of Sciences, Dalian 116023, China

Abstract Gas-liquid microreaction technology has shown great potential in a variety of industrial relevant mass transfer operations and reactions. This paper outlines the current research status of this technology with emphasis on reactor design, hydrodynamics and mass transfer phenomena as well as reaction applications. The future challenges of this important technology are also summarized.
Keywords microchannel, microreactor, two-phase flow, flow pattern, gas-liquid reaction

1 INTRODUCTION

Nowadays, one of the major challenges that the chemical industry has to cope with is to create innovative processes with less pollution, improved chemistry and high energy efficiency [1]. Microchemical technology provides unique solutions to meet these requirements. As an important means for process intensification, microchemical technology is expected to have a number of advantages for chemical related production [2]. The high heat and mass transfer rates in microchemical systems enable a lot of highly exothermic, fast reactions to be operated under nearly isothermal conditions, thereby better selectivity or yield can be reached as compared to conventional reactors. Also the small reactant inventories of microchemical systems lead to better process safety, which especially favors the on-site distributed production of extremely toxic or hazardous substances. Moreover, microchemical technology adopts a numbering-up approach to increase the system throughput (*i.e.*, through the replication of the functional microstructured unit) so that the scale-up becomes relatively easy. This further increases the production flexibility in response to varying market demands.

The last decade has witnessed a rapid development in microchemical technology. A scientific conference series of "International Conference on Microreaction Technology" has been held yearly since 1997, which aims at the communication of the newest development of microchemical technology among industrial companies and research institutions all over the world. Now it is commonly acknowledged that microchemical engineering has become an independent discipline in chemical and process engineering [3]. As an important field of this technology, gas-liquid microreaction technology has attracted increased attention in recent years due to its great potential in many industrial important gas-liquid mass transfer operations and chemical reactions [2–4]. This article outlines the current research status of gas-liquid microreaction technology. The main topics include reactor design, hydrodynamics, mass transfer and some reaction applications. The future development of this technology and some challenges are also discussed.

2 GAS-LIQUID MICROREACTOR DESIGN

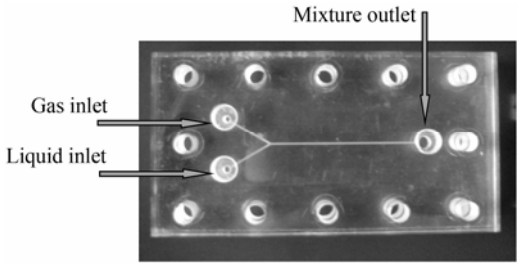
Figure 1 shows some typical photographs of the available designs of gas-liquid microreactor. According to mechanism of contact between two phases, gas-liquid microreactor can be mainly classified into four categories [4]:

Microchannel contactor Gas and liquid flow concurrently in the same microchannel where different flow patterns can be established. The simplest microchannel contactor comprises only one microchannel for two-phase contact [5] (called single microchannel contactor), as shown in Fig. 1 (a). A parallel microchannel contactor consists of a multitude of microchannels connected in parallel. In the latter case the design of inlet fluid distributors becomes critical in order to ensure a uniform distribution of two phases among microchannels. Different design criteria have been conceived in the literature, such as large buffer volume design [6], separate inlet high pressure drop channels [7, 8] and constructal distributors [9] [see Fig. 1 (b)].

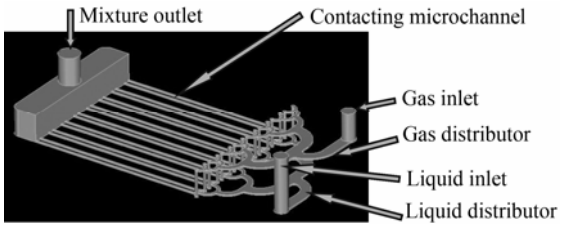
Falling film microreactor The falling film microreactor utilizes the gravity force to generate thin liquid films in microchannels. A well known falling film microreactor developed by Institut für Mikrotechnik Mainz (IMM) is shown in Fig. 1 (c) [7]. The central part of this microreactor is a stainless steel plate containing 64 vertically positioned microchannels (300 μm wide and 100 μm deep). Liquid spreads to form a thin film among microchannels and flows further downward to the withdrawal zone at the bottom. Gas flows in a large gas chamber positioned above the microchannel section, facilitating concurrent or countercurrent operation mode.

Microreactor with phase interface stabilized by physical structures Gas and liquid flow in two adjacent microchannels which are separated by physical structures such as meshes [10], micro-porous plates [11] or overlapping channel configurations [12]. Thus two-phase interface can be formed and stabilized in

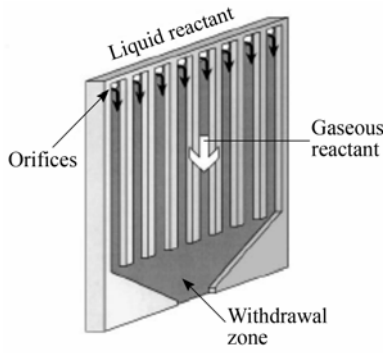
Received 2008-07-08, accepted 2008-08-18.
* Supported by the National Natural Science Foundation of China (20490208, 20676129), the National High Technology Research and Development Program of China (2006AA05Z233, 2007AA030206).
** To whom correspondence should be addressed. E-mail: qyuan@dicp.ac.cn



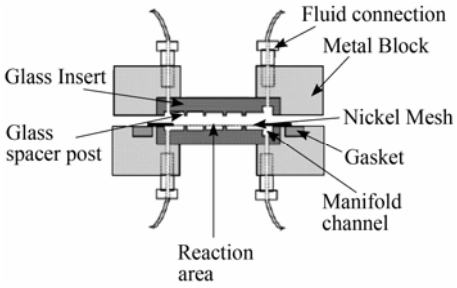
(a) Single microchannel contactor [5]



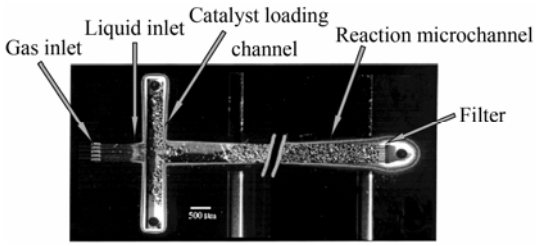
(b) Parallel microchannel contactor [9]



(c) Falling film microreactor [7]



(d) Mesh microreactor [10]



(e) Micro packed bed reactor [13]

Figure 1 Different types of gas-liquid microreactor

the well-defined openings of these structures. Fig. 1 (d) shows a mesh microreactor designed by Wenn *et al*. [10]. The reactor consists of a nickel mesh positioned between two glass inserts in order to form two separate fluid chambers. The depth of each chamber is set at 100 μm. Gas-liquid contact is realized at arrays of openings microfabricated in the mesh. The flow arrangement can be concurrent, countercurrent or cross flow.

Micro packed bed reactor When solid catalysts are involved in gas-liquid reactions performed in the three types of microreactor mentioned above, they can be integrated as wall coatings through methods such as sol-gel techniques, and anodic oxidation [2]. The micro packed bed reactor incorporates catalyst particles directly in microchannels. Fig. 1 (e) shows a possible design [13]: gas and liquid form a multilamination flow arrangement before they meet at the entrance of the reaction microchannel (625 μm wide and 20 cm long). Another 400 μm wide microchannel perpendicular to the reaction microchannel is used to delivery the catalyst slurry. At the outlet of the reaction microchannel, a microfilter comprising an array of posts was etched in order to retain the packed catalyst. To further increase the throughput of this microreactor, a multichannel design should be adopted.

Among the above four designs of gas-liquid microreactor, microchannel contactor seems to be the most attractive one for industrial applications in view of the fact that it can offer relatively high mass transfer capacity and good reaction efficiency under a wide range of flow rates of gas and liquid [14]. This is exactly the case when gas-liquid reactions without catalysts involved are concerned. With regard to gas-liquid-solid catalyzed reactions, finding some possible ways to immobilize highly active catalyst coatings onto the microchannel wall is a necessity for the successful application of such microreactor. The micro packed bed reactor concept provides another option, that is, the direct incorporation of catalyst pellets ready available inside microchannels might be more preferable. In contrast, the other two types of microreactor may find some uses in applications where a fast gas-liquid separation after reaction or a very low pressure drop across the reactor must be guaranteed.

3 HYDRODYNAMICS AND MASS TRANSFER

A thorough understanding of hydrodynamics and mass transfer characteristics in gas-liquid microreactors is of vital importance to the prediction of the reactor performance.

3.1 Microchannel contactor

3.1.1 *Gas-liquid two-phase flow in a single microchannel*

For concurrent gas-liquid flow in one microchannel, there exist different flow patterns depending on the values of superficial gas and liquid velocities (*i.e.*, u_G and u_L). Many flow pattern visualization experiments have been conducted in circular, rectangular and even triangular microchannels [14–19]. It is demonstrated that the flow pattern of CO_2-water flow in a rectangular microchannel with cross section of 500 μm×1000 μm includes bubbly flow, slug flow (composed of two sub-regimes: Taylor flow and unstable slug flow), slug-annular flow, churn flow and annular flow [18] as shown in Fig. 2. The disappearance of stratified flow implies that the role of buoyancy is largely suppressed in microchannels. The available studies also reveal that due to the importance of surface tension and laminar flow nature in microchannels, the widely used flow transition correlations or models in large channels generally cannot be applied to microchannels. Towards solving this problem, Triplett *et al*. [15] and Akbar *et al*. [20] have proposed two representative flow pattern maps based on coordinates of phasic superficial velocities and Weber numbers, respectively. However, it has been found [18] that the two flow pattern maps can only describe flow transition behavior in large microchannels with diameters close to 1 mm (see Fig. 2). This necessitates more work on the pursuit of reasonable flow transition models for microchannels. Under operational conditions relevant to gas-liquid reactions in microchannel contactors, Taylor flow is the preferred flow pattern due to its advantageous flow properties, *e.g.*, the separation of the bulk liquid by bubbles indicates low liquid axial mixing and the inner circulation in the liquid slug region further improves liquid radial mixing.

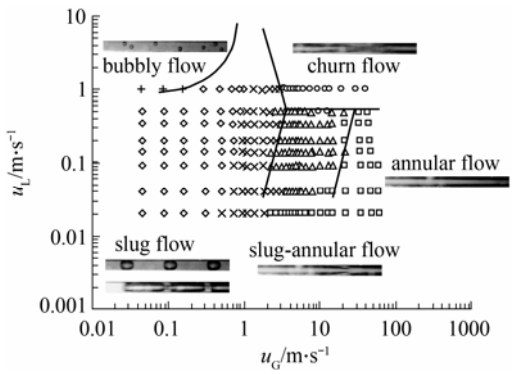


Figure 2 Flow pattern map of CO_2-water flow in a rectangular microchannel with a cross section of 500 μm×1000 μm [18]
+bubbly flow; ◇Taylor flow; × unstable slug flow; △slug-annular flow; □annular flow; ○churn flow; —— Triplett *et al.* [15]

Pressure drop is another important parameter associated with gas-liquid flow in microchannels. The pressure drop correlations based on the traditional homogeneous flow model and separated flow model were found to have limited applicability in microchannels [16, 21, 22]. In bubbly flow, the bubbles are randomly dispersed in liquid with sizes much smaller than the channel diameter. Thus two phases can be treated as a pseudo-fluid with mixture properties, implying the validity of the homogeneous flow model [16, 21]. In flow patterns including annular flow, slug-annular flow and churn flow, a nearly continuous gas flow is seen in the center part of the microchannel while the liquid film flows adjacent to the microchannel wall, which is close to the assumptions of the separated flow model. Our studies have demonstrated [18] that the well-known Lockhart and Martinelli correlation based on this model can still represent pressure drop data in these flow patterns in rectangular microchannels if some appropriate modifications have been made to this correlation to account for the observed mass flux effect. For Taylor flow, a more reasonable model should express two-phase frictional pressure drop as a sum of the respective frictional loss in the liquid slug, in the liquid film and pressure drop over the bubble cap [23]. Thus the inner flow details of this flow pattern are indispensable for its pressure drop prediction.

3.1.2 *Two-phase flow distribution in parallel microchannels*

In parallel microchannel contactors, two-phase flow uniformity depends crucially on the inlet gas-liquid distributing principle as well as the ideal flow pattern involved (*i.e.*, that under two-phase flow equipartition). De Mas *et al*. [24] claimed that the dynamic pressure fluctuation associated with slug flow in parallel microchannels would lead to two-phase flow instability and eventually to channeling if the flow resistance through gas and liquid inlets was low. To operate parallel microchannel contactors under uniform slug flow distribution, two phases should be introduced *via* individual inlet channels that present a pressure drop significantly higher than that across the subsequent contacting microchannels. This rule of thumb has been verified by recent experiments [8]. When the ideal flow pattern shifts to slug-annular flow or annular flow, the dynamic pressure fluctuation is reduced to a great extent so that two-phase flow therein can be essentially treated as steady. In this case, the arrangement of inlet high pressure drop channels is no longer a requisite whereas many manifold designs widely used for single-phase flow equipartition can be directly employed. For example, gas or liquid can be introduced into each microchannel through a buffer reservoir that has a volume significantly larger than that of parallel microchannels [6]. Alternatively, a more efficient design can be envisaged from the integration of two constructal distributors with dichotomic tree structures in front of parallel microchannels [9], as shown in Fig. 1 (b).

The production capacity of a microchanel contactor can be further increased by stacking more reaction platelets comprising parallel microchannels together, which highly relies on the availability of fast and cost-effective large-scale microfabrication techniques as well as assembly methods of microreactors. Regarding industrial applications, the parallel setup of numerous microreactors should also be envisaged in order to afford the desired production rate. In this case, uniform two-phase flow distribution in the whole microchemical system presents an even bigger challenge.

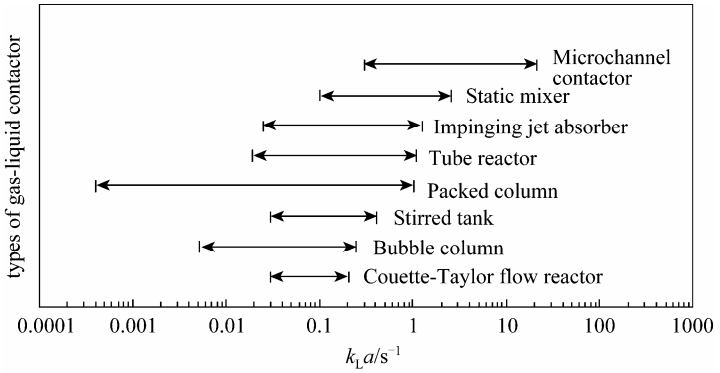
3.1.3 *Gas-liquid mass transfer*

Figure 3 shows the comparison of mass transfer parameters measured in a rectangular microchannel having cross-section of 500 μm×1000 μm with other types of gas-liquid contactors [14]. It is seen that the liquid side volumetric mass transfer coefficient, k_La, and interfacial area, a, in this microchannel can reach 21 s^{-1} and 9000 $m^2 \cdot m^{-3}$, respectively. These values exceed those in conventional contactors such as bubble columns, and packed columns by at least one or two orders of magnitude. To elucidate mass transfer intensification mechanism in microchannels, an investigation of mass transfer details inside each flow pattern is necessary. In view of significant advantages of Taylor flow for gas-liquid reactions in microchannels, most mass transfer studies are devoted to this flow pattern [9, 25, 26]. The results suggest that the transfer of gas can be realized *via* the interface between two bubble caps and the touching liquid slug or *via* the interface between the bubble body and the surrounding liquid film. Mass transfer contributions through these two types of interface are mainly determined by the contact time between the liquid film and the bubble, the mixing extent of the liquid film to the liquid slug, and the Taylor bubble length fraction. Consequently, the overall k_La highly depends on the inner flow parameters. At large values of bubble velocity and liquid slug length which correspond to short film contact time and complete mixing of the liquid film into the liquid slug, van Baten *et al*. [25] showed *via* Computational Fluid Dynamics (CFD) simulation that k_La values can be described by their mass transfer model in which the contributions through the two interfaces were both derived based on the penetration theory. For other mass transfer circumstances (*e.g*., long film contact time or poor mixing of the film into the liquid slug), different empirical correlations have also been proposed [9, 26]. However, the relevant theoretic insights are still not well clarified, which requires further numerical analysis as well as experimental validation. Besides Taylor flow, annular flow is another attractive flow pattern because of the extremely high interfacial area it provides [2]. However, a pressure drop penalty is also imposed since this flow pattern occurs under very high superficial gas velocities (see Fig. 2).

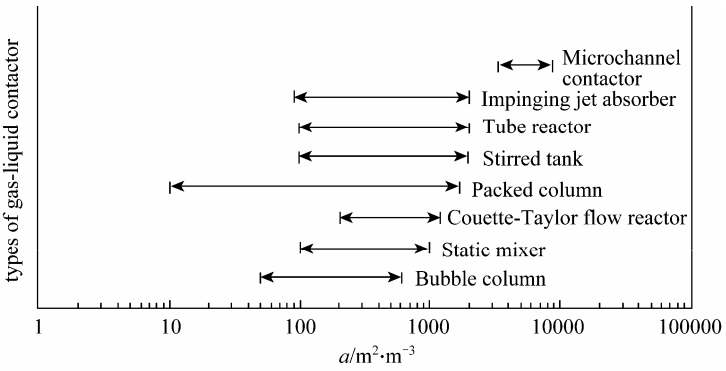
3.2 Falling film microreactor

3.2.1 *Falling film hydrodynamics in microchannels*

The flow of the falling liquid film in microchannels differs significantly from that on conventional flat plates. As a result of small channel sizes and the action of capillary forces, the surface of the liquid film in microchannels usually takes the form of a flowing meniscus rather than completely flat film. Meanwhile, the flow of the falling film in microchannels should be treated as a three-dimensional flow and the increased importance of surface tension, evaporation, surface heterogeneity and roughness characteristics should be well addressed. Consequently, the existing equations and correlations from the literature cannot be applied to predict the average thickness of the falling film in microchannels [27].



(a) Liquid side volumetric mass transfer coefficient



(b) Interfacial area

Figure 3 Comparison of mass transfer performance in different gas-liquid contactors [14]

3.2.2 *Gas flow through the gas chamber*

Commenge *et al*. [28] conducted both experimental and numerical analysis of gas phase residence time distribution (RTD) in a commercially available falling film microreactor as that shown in Fig. 1 (c). Considerable back-mixing in the gas phase was found when the gas Reynolds number was higher than 24 due to the presence of two hydrodynamic effects at the entrance of the gas chamber (the formation of recirculation loops and the jet effect), which created a wide RTD spread. Thus the design concerning the gas feed in the falling film microreactor of this type should be further improved.

3.2.3 *Mass transfer characteristics*

The falling film microreactor can generate stable films less than 100 μm thick. Thus the diffusion path in the liquid phase is greatly shortened and very high interfacial area can be offered, which substantially improves the gas-liquid mass transfer process. The formulation of a reasonable gas-liquid mass transfer model for the falling film microreactor requires that three-dimensional equations of momentum, mass, and energy balances (if heat effects are involved) be solved together, which is rather difficult due to the complex falling film hydrodynamics in microchannels. As a first approximation, a steady one-dimensional flow of the liquid film with a flat surface profile can be assumed. Then the average thickness of the liquid film and its velocity distribution can be easily determined. Thus, the derivation of mass transfer parameters becomes a relatively simple task [29].

3.3 Microreactor with phase interface stabilized by physical structures

In this type of microreactor, single-phase flows of gas and liquid are seen in two adjacent microchannels that are separated by a physical structure with well-defined openings. In order to form a stabilized interface inside the openings and to prevent the breakthrough of one phase into the other, the flow rates of gas and liquid should be constrained by a relatively low pressure difference between two phases. The allowable maximum pressure difference can be estimated from the Laplace equation on the capillary pressure.

Such microreactor allows the mass transfer between gas and liquid in parallel flow configuration without the occurrence of phase mixing. To ensure a sufficiently low diffusive resistance in each phase, the depth of respective microchannels should be held as small as possible. Moreover, two-phase contact region in the additional microstructure separating two microchannels requires proper design so that the diffusive resistance therein will be lowered significantly as well. A value of interfacial area per liquid volume of about 2000 $m^2 \cdot m^{-3}$ has been reported for the mesh microreactor shown in Fig. 1 (d) [30], demonstrating its good mass transfer efficiency. However, mass transfer capacity in this sort of microreactor is somewhat limited as compared to microchannel contactors, in view of the fact that in the former case gas is transferred into liquid predominantly by molecular diffusion.

3.4 Micro packed bed reactor

Gas-liquid flow in the micro packed bed reactor does not show much difference from that observed in its macroscale counterpart. For example, flow regimes of hydrogen-cyclohexene mixture in the micro packed bed reactor illustrated in Fig. 1 (e) were found to include bubble flow, steady concurrent flow and pulsating flow depending on the values of the flow rates of gas and liquid [13], which was in accordance with the description of flow regimes previously used for traditional trickle bed reactors.

Due to the small channel dimensions in the micro packed bed reactor, the size of catalyst particles has to be very small (typically lower than several tens micrometers). Thus prohibitively high gas-liquid interfacial area can be generated, which directly translates into an enhancement in gas-liquid mass transfer rate [13].

4 SOME REACTION APPLICATIONS

4.1 Direct fluorination

The direct fluorination using elemental fluorine is usually difficult to proceed in conventional reactors due to the presence of explosive risks and large quantities of side products. Gas-liquid microreaction technology will find wide uses in such reactions *via* efficient thermal management and better control of gas-liquid mixing. The technological benefits of performing the direct fluorination of toluene in microreactors have been well demonstrated [7, 24]. Yield of monofluorinated toluene of up to 28% was reported in a falling film microreactor by Jähnisch *et al*. [7]. The space-time yields in this microreactor were orders of magnitude higher than those achieved in a laboratory bubble column. Besides toluene, the direct fluorination of various other organic compounds including 4-nitrotoluene, 1,3-dicarbonyl substrates have been carried out successfully in microchannel contactors [6, 31]. The reactions proceeded efficiently under annular flow. In the case of fluorination of ethyl-2-chloro-3- oxobutanoate in a triple microchannel contactor, the substrate conversion could reach 59% and the amount of the monofluorinated species in the crude product after solvent removal was 74%, which compares favorably with the amount of 85% of the monofluorinated species in the product observed in a laboratory stirred vessel when the conversion was 15% [31, 32].

4.2 Liquid phase hydrogenation

Liquid phase hydrogenation represents an important classification of gas-liquid reactions ubiquitous in the industry. To enable the fast transfer of hydrogen into the liquid, reactions are usually operated under high pressures in conventional reactors. The enhanced mass transfer rate in microreactors suggests that they are good alternative reactors, where promising conversions or yields can be reached. For example, Kobayashi *et al*. [33] investigated the hydrogenation of benzalacetone over a Pd catalyst in a single microchannel

contactor (200 μm wide, 100 μm deep and 45 cm long). The yield of 4-phenyl-2-butanone could reach 97% when operated under annular flow at a liquid space velocity of about 11 h^{-1} (based on the microchannel volume). The microreactor functioned well during continuous operation of 5 h. Nevertheless, the long-term stability of the immobilized Pd catalyst still remains unclear.

4.3 Direct synthesis of hydrogen peroxide

Inoue *et al*. [34] have performed hydrogen peroxide synthesis by the direct combination of hydrogen and oxygen over palladium catalysts in a 10-channel micro packed bed reactor. The reaction proceeded safely in the reactor even at hydrogen/oxygen ratios in the explosive regime (*e.g*., up to 1:1) under pressures of 2−3 MPa [the explosion limits for hydrogen in oxygen range from about 4% to 94% (by volume)]. Meanwhile, the transfer rate of hydrogen and oxygen to the liquid was significantly enhanced in the micro packed bed reactor compared to conventional reactors, which made it possible to obtain a faster reaction rate.

4.4 Chemical absorption

Chemical absorption is widely used in chemical plants, *e.g*., for the purification of exhaust gases. Due to relatively lower mass transfer rate in the existing absorption apparatus, they usually occupy large space. In contrast, substantially improved mass transfer process in gas-liquid microreactors implies that they can provide much higher absorption efficiency while being more compact. As an example, promising results have been seen in the work of TeGrotenhuis *et al*. [11] who performed gas absorption experiments by using a microreactor in which gas and liquid were separated by a micro-porous plate. It was found that over 90% of CO_2 could be removed from the mixture stream containing 25% CO_2 in less than 10 s gaseous residence time when using diethanolamine solution as the absorbent.

4.5 Engineering projects

Although most research efforts of gas-liquid microreaction technology still remain on the bench scale, it is noteworthy that a few industrial companies in Europe and the United States have launched some engineering projects dedicated to the commercialization of this technology [35, 36]. Among them, UOP is ambitious to construct a microreactor plant for the production of hydrogen peroxide in the order of about 150000 metric tons per year *via* the direct combination of hydrogen and oxygen in a micro packed bed reactor [36]. A laboratory test at IMM verified that a space-time yield of 2 g hydrogen peroxide per g catalyst h could be achieved at a pressure of 3 MPa and hydrogen/oxygen ratio of 1∶3. UOP then carried out pilot-scale tests at higher pressures to study the reaction kinetics and conditions and to analyze the capital costs. The full-scale plant is said to be built in the forthcoming few years.

5 FUTURE CHALLENGES

Gas-liquid microreaction technology is now making a rapid progress in many fields, however, its implementation in the industry still faces a number of challenges, as summarized below.

Fluid mechanics aspect At present, our understanding of multiphase flow characteristics in microreactors is mostly experiment-based. The relevant theoretical bases should be established. As far as the existing types of gas-liquid microreactors are concerned (see Fig. 1), fundamental topics to be tackled include the mechanism of formation of gas-liquid two-phase flow pattern and its transition rule in a single microchannel, gas-liquid two-phase flow instability in parallel microchannels, motion characteristics of falling liquid film in microchannels, interfacial phenomena in microfabricated openings, and mechanics of gas-liquid flow through micrometer-sized irregular void space. The former two topics should be paid more attention in view of the promising application potential of microchannel contactors. Research highlights are most likely expected in two-phase fluid mechanics studies in microchannels through the use of novel measurement techniques such as microscopic particle image velocimetry (μ-PIV) [37]. Besides experimental study, numerical techniques such as CFD simulations and lattice Boltzmann method also open up new ways [38, 39].

Reactor model The design and operation of gas-liquid microreactors requires comprehensive reactor models that can well represent the interplay between transport phenomena and chemical kinetics therein. Such models are not available as yet, primarily due to complex multiphase flow characteristics in microreactors, although some preliminary efforts have been seen [29, 34, 40]. The formulation of reactor models that can provide a predictive description of heat and mass transfer effects, and chemical reactions under different gas-liquid flow patterns in microreactors is therefore a must in the near term. For Taylor flow in microchannel contactors, the relevant models might be easily derived based on the existing knowledge of two-phase reaction study under this flow pattern in millimeter-sized channels that has been widely explored over the past decades [41].

Numbering-up issues The numbering-up strategy brings several research and technical challenges. Proper system integration schemes should be first devised in order to assemble many microreactors together and to facilitate the incorporation of auxiliary reaction control, and monitoring components such as heat exchangers and chemical sensors. Some possible arrangements have been suggested [42]. Handling of fluids in such enlarged microchemical systems also requires multiscale structure designs so that the connection from the microscale channel structures all the way to the macroscale pipe networks can be optimized. The constructal approach seems to address this issue very well [43]. Moreover, technical solutions to some

important problems that may arise in practical applications are not available as yet, *e.g*., the preparation of active catalyst coatings with long-term mechanical and chemical stability, and the avoidance of blockage in microreactors.

To put gas-liquid microreaction technology into industrial practices requires extensive efforts among industrial partners and academic institutions. Some standard gas-liquid microreactor components are already commercially available. Days are thus not far away when a wide application of gas-liquid microreaction technology comes true. This will radically change the appearance of the existing chemical plants. Notably, the future plants will be much smaller, safer and cleaner.

REFERENCES

1 Charpentier, J.C., "The triplet 'molecular processes-product-process' engineering: the future of chemical engineering?", *Chem. Eng. Sci*., **57**, 4667−4690 (2002).
2 Ehrfeld, W., Hessel, V., Löwe, H., Microreactors: New Technology for Modern Chemistry, Wiley-VCH, Weinheim (2000).
3 Hessel, V., Hardt, S., Löwe, H., Chemical Micro Process Engineering: Fundamentals, Modelling and Reactions, Wiley-VCH, Weinheim (2004).
4 Hessel, V., Angeli, P., Gavriilidis, A., Löwe, H., "Gas-liquid and gas-liquid-solid microstructured reactors: contacting principles and applications", *Ind. Eng. Chem. Res*., **44**, 9750−9769 (2005).
5 Yue, J., Chen, G., Yuan, Q., Luo, L., Le Gall, H., "Mass transfer in gas-liquid flow in microchannels", *J. Chem. Ind. Eng.* (*China*), **57**, 1296−1303 (2006). (in Chinese)
6 Chambers, R.D., Fox, M.A., Holling, D., Nakano, T., Okazoeb, T., Sandford, G., "Elemental fluorine. Part 16. Versatile thin-film gas-liquid multi-channel microreactors for effective scale-out", *Lab Chip*, **5**, 191−198 (2005).
7 Jähnisch, K., Baerns, M., Hessel, V., Ehrfeld, W., Haverkamp, V., Löwe, H., Wille, Ch., Guber, A., "Direct fluorination of toluene using elemental fluorine in gas/liquid microreactors", *J. Fluorine Chem*., **105**, 117−128 (2000).
8 de Mas, N., Günther, A., Kraus, T., Schmidt, M.A., Jensen, K.F., "Scale-out multilayer gas-liquid microreactor with integrated velocimetry sensors", *Ind. Eng. Chem. Res*., **44**, 8997−9013 (2005).
9 Yue, J., "Hydrodynamics and mass transfer characteristics in gas-liquid microchannel contactors", Ph.D. Thesis, Dalian Institute of Chemical Physics, Chinese Academy of Sciences, Dalian (2008). (in Chinese)
10 Wenn, D.A., Shaw, J.E.A., Mackenzie, B., "A mesh microreactor for 2-phase reactions", *Lab Chip*, **3**, 180−186 (2003).
11 TeGrotenhuis, W.E., Cameron, R.J., Viswanathan, V.V., Wegeng, R.S., "Solvent extraction and gas absorption using microchannel contactors", In: Proceedings of the Third International Conference on Microreaction Technology, Springer Verlag, Berling, 541−549 (2000).
12 Hibara, A., Iwayama, S., Matsuoka, S., Ueno, M., Kikutani, Y., Tokeshi, M., Kitamori, T., "Surface modification method of microchannels for gas-liquid two-phase flow in microchips", *Anal. Chem*., **77**, 943−947 (2005).
13 Losey, M.W., Schmidt, M.A., Jensen, K.F., "Microfabricated multiphase packed-bed reactors: characterization of mass transfer and reactions", *Ind. Eng. Chem. Res*., **40**, 2555−2562 (2001).
14 Yue, J., Chen, G., Yuan, Q., Luo, L., Gonthier, Y., "Hydrodynamics and mass transfer characteristics in gas-liquid flow through a rectangular microchannel", *Chem. Eng. Sci*., **62**, 2096−2108 (2007).
15 Triplett, K.A., Ghiaasiaan, S.M., Abdel-Khalik, S.I., Sadowski, D.L., "Gas-liquid two-phase flow in microchannels. Part I: Two-phase flow patterns", *Int. J. Multiphase Flow*, **25**, 377−394 (1999).
16 Chung, P.M.-Y., Kawaji, M., "The effect of channel diameter on adiabatic two-phase flow characteristics in microchannels", *Int. J. Multiphase Flow*, **30**, 735−761 (2004).
17 Cubaud, T., Ho, C-M., "Transport of bubbles in square microchannels", *Phys. Fluids*, **16**, 4575−4585 (2004).
18 Yue, J., Luo, L., Gonthier, Y., Chen, G., Yuan, Q., "An experimental investigation of gas-liquid two-phase flow in single microchannel contactors", *Chem. Eng. Sci*., **63**, 4189−4202 (2008).
19 Zhao, T.S., Bi, Q.C., "Co-current air-water two-phase flow patterns in vertical triangular microchannels", *Int. J. Multiphase Flow*, **27**, 765−782 (2001).
20 Akbar, M.K., Plummer, D.A., Ghiaasiaan, S.M., "On gas-liquid two-phase flow regimes in microchannels", *Int. J. Multiphase Flow*, **29**, 855−865 (2003).
21 Triplett, K.A., Ghiaasiaan, S.M., Abdel-Khalik, S.I., Lemouel, A., McCord, B.N., "Gas-liquid two-phase flow in microchannels. Part II: Void fraction and pressure drop", *Int. J. Multiphase Flow*, **25**, 395−410 (1999).
22 Yue, J., Chen, G., Yuan, Q., "Pressure drops of single and two-phase flows through T-type microchannel mixers", *Chem. Eng. J*., **102**, 11−24 (2004).
23 Kreutzer, M.T., Kapteijn, F., Moulijn, J.A., Kleijn, C.R., Heiszwolf, J., "Inertial and interfacial effects on pressure drop of Taylor flow in capillaries", *AIChE J*., **51**, 2428−2440 (2005).
24 de Mas, N., Gunther, A., Schmidt, M.A., Jensen, K.F., "Microfabricated multiphase reactors for the selective direct fluorination of aromatics", *Ind. Eng. Chem. Res*., **42**, 698−710 (2003).
25 van Baten, J.M., Krishna, R., "CFD simulations of mass transfer from Taylor bubbles rising in circular capillaries", *Chem. Eng. Sci*., **59**, 2535−2545 (2004).
26 Berčič, G., Pintar, A., "The role of gas bubbles and liquid slug lengths on mass transport in the Taylor flow through capillaries", *Chem. Eng. Sci*., **52**, 3709−3719 (1997).
27 Yeong, K.K., Gavriilidis, A., Zapf, R., Hessel, V., "Experimental studies of nitrobenzene hydrogenation in a microstructured falling film reactor", *Chem. Eng. Sci*., **59**, 3491−3494 (2004).
28 Commenge, J.M., Obein, T., Genin, G., Framboisier, X., Rode, S., Schanen, V., Pitiot, P., Matlosz, M., "Gas-phase residence time distribution in a falling-film microreactor", *Chem. Eng. Sci*., **61**, 597−604 (2006).
29 Zanfir, M., Gavriilidis, A., Wille, Ch., Hessel, V., "Carbon dioxide absorption in a falling film microstructured reactors: experiments and modeling", *Ind. Eng. Chem. Res*., **44**, 1742−1751 (2005).
30 Abdallah, R., Meille, V., Shaw, J., Wenn, D., de Bellefon, C., "Gas-liquid and gas-liquid-solid catalysis in a mesh microreactor", *Chem. Commun*., **4**, 372−373 (2004).
31 Chambers, R.D., Holling, D., Spink, R.C.H., Sandford, G., "Elemental fluorine. Part 13. Gas-liquid thin film microreactors for selective direct fluorination", *Lab Chip*, **1**, 132−137 (2001).
32 Chambers, R.D., Greenhall, M.P., Hutchinshon, J., "Direct fluorination of 1,3-dicarbonyl compounds", *Tetrahedron*, **52**, 1−8 (1996).
33 Kobayashi, J., Mori, Y., Okamoto, K., Akiyama, R., Ueno, M., Kitamori, T., Kobayashi, S., "A microfluidic device for conducting gas-liquid-solid hydrogenation reactions", *Science*, **304**, 1305−1308 (2004).
34 Inoue, T., Schmidt, M.A., Jensen, K.F., "Microfabricated multiphase reactors for the direct synthesis of hydrogen peroxide from hydrogen and oxygen", *Ind. Eng. Chem. Res*., **46**, 1153−1160 (2007).
35 Hessel, V., Löwe, H., Müller, A., Kolb, G., Chemical Micro Process Engineering: Processing and Plants, WILEY-VCH, Weinheim (2005).
36 Pennemann, H., Hessel, V., Löwe, H., "Chemical microprocess technology-from laboratory-scale to production", *Chem. Eng. Sci*., **59**, 4789−4794 (2004).
37 van Steijn, V., Kreutzer, M.T., Kleijn, C.R., "μ-PIV study of the formation of segmented flow in microfluidic T-junctions", *Chem. Eng. Sci*., **62**, 7505−7514 (2007).
38 Hardt, S., Doffing, F., Pennemann, H., "Simulation of hydrodynamic dispersion in gas/liquid microreactors", In: Proceedings of the Fifth Conference on Modeling and Simulation of Microsystems, 22−25 April, San Juan, Puerto Rico, 54−57 (2002).
39 Yu, Z., Hemminger, O., Fan, L.S., "Experiment and lattice Boltzmann simulation of two-phase gas-liquid flows in microchannels", *Chem. Eng. Sci*., **62**, 7172−7183 (2007).
40 Haverkamp, V., Hessel, V., Liauw, M.A., Löwe, H., Menges, M.G., "Reactor model for fast reactions in the micro-bubble column and validation", *Ind. Eng. Chem. Res*., **46**, 8558−8565 (2007).
41 Roy, S., Bauer, T., Al-Dahhan, M., Lehner, P., Turek, T., "Monoliths as multiphase reactors: A review", *AIChE J*., **50**, 2918−2934 (2004).
42 Jensen, K.F., "Microreaction engineering - is small better?", *Chem. Eng. Sci*., **56**, 293−303 (2001).
43 Luo, L., Tondeur, D., Le Gall, H., Corbel, S., "Constructal approach and multi-scale components", *Appl. Therm. Eng*., **27**, 1708−1714 (2007).